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Abstract

PREPARATION AND PHYSICAL PROPERTIES OF UO_2 SINGLE CRYSTALS

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An extensive study of different fundamental physical properties of UO_2 single crystals of different compositions has been undertaken. The different properties were studied before and after reactor irradiation. This work includes the preparation of single crystals, the variation of composition of the single crystals by controlled oxidation, sintering of single crystal spheres, transmission electron microscope observation of thin foils and also the measurement of the thermal conductivity, the electrical conductivity, the thermoelectric power, the optical absorption spectrum, the elastic constants and of the resonant γ-ray absorption.

The results obtained as yet are reviewed in this communication. A method for growing uranium dioxide single crystals by sublimation has been developed. High quality semi-spherical crystals from 4 to 15 mm diameter, up to 7 g in weight, can be prepared. The stoichiometric oxygen excess of the UO_{2+x} crystals can afterwards be varied in the range $0 < x < 0.25$ by carefully controlled oxidation. This was achieved by a new, simple and reliable method.

UO_2 single crystals can be prepared as thin foils suitable for transmission electron microscopic observation. The thinning is achieved chemically at 200°C or electrolytically at room temperature. Dislocations, precipitates of different shapes, consisting presumably of U_4O_9, fission fragment damage, as well as tracks, clusters of point defects and small dislocation loops, are observed and discussed.

It has been possible to cut square elongated prisms of about 8 mm in length from the vapour grown single crystals. These prisms are suitable for the determination of the thermal conductivity. The latter has been measured from liquid helium temperature up to room temperature. No striking qualitative deviation has been found from the results obtained by other investigators using sintered polycrystalline material.

The electrical conductivity and the thermoelectric power of UO_2 single crystals of different composition have been measured in an extended temperature range before and after reactor irradiation. The latter results in a very marked decrease of the conductivity of the non-stoichiometric crystals. This effect can be attributed to the formation of hole traps during irradiation. From recovery experiments, one concludes to the existence of an annealing peak around 725°C, the associated activation energy being about 2.5 eV. This suggests possible uranium point defect migration.

From the analysis of the optical absorption spectrum, a value of 2.18 eV is found for the absorption edge at room temperature.

The elastic constant has been determined using a pulse echo technique, which allows a precision of about 1% .

UO_2 doped with Fe^{57} shows an unsplit single peak Mössbauer spectrum. However, if kept for nine months at room temperature a spectrum appears which can be analysed as arising from the superposition of this peak and those arising from magnetic hyperfine splitting.